Research Progress on Microbial Fuel Cell Systems for Treating Antibiotic Wastewater

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Abstract

The widespread use of antibiotics in biomedical fields has led to their substantial residues in the environment, and the resulting bacterial resistance has become a global health threat. Conventional antibiotic wastewater treatment processes have limited capacity for antibiotic removal. As an emerging bio electrochemical technology, microbial fuel cells (MFCs) can degrade antibiotics while simultaneously recovering electrical energy, demonstrating broad prospects in the field of antibiotic wastewater treatment. This review comprehensively introduces and summarizes the degradation performance and electricity generation of MFCs toward different types of antibiotics, the treatment of antibiotic wastewater by various biological processes, and discusses the advantages and challenges of coupled MFC systems in synergistically enhancing degradation, reducing antibiotic resistance genes, and improving electricity generation performance. This paper aims to provide a reference for in-depth research and practical application of MFCs and their coupled technologies in antibiotic wastewater treatment.

Keywords

Microbial fuel cells, Antibiotic wastewater, Antibiotic removal, Coupled system

Introduction

Antibiotics, owing to their effectiveness in inhibiting bacterial infections, are extensively used in human disease treatment, livestock husbandry, and aquaculture. However, antibiotics are incompletely metabolized in organisms. Large amounts of parent compounds enter the environment via excreta, continuously accumulate, and induce bacterial resistance and corresponding resistance genes, posing a serious threat to human health. According to statistics, the global consumption of antibiotics exceeded 90,000 tons in 2017 and is projected to reach 100,000 tons by 2030, with the consumption for human disease treatment expected to increase by 15.0% compared with 2017. As one of the world's largest producers and consumers of antibiotics, China faces an urgent challenge in addressing the environmental pollution caused by antibiotics [1-4].

It is estimated that by 2050, the annual death toll from drug-resistant pathogens will reach 10 million, surpassing the current number of cancer-related deaths. Wastewater treatment plants are among the major anthropogenic sources of antibiotics, antibiotic resistance genes (ARGs), and antibiotic-resistant bacteria (ARB) released into the environment [5,6]. However, conventional wastewater treatment processes have limited capacity to remove antibiotics, and low concentrations of antibiotics can persist in the aquatic environment [7]. Traditional antibiotic wastewater treatment technologies mainly include physical adsorption, advanced oxidation, membrane separation, and conventional biological treatment. Nevertheless, these technologies have certain limitations, such as high cost, low efficiency, and low throughput. They have limited mineralization ability, may generate toxic oxidation intermediates, and can potentially introduce ARGs into microorganisms, thereby producing more harmful by-products [8]. Consequently, developing efficient, economical, and sustainable antibiotic wastewater treatment technologies has become a research focus in the environmental field.

Microbial fuel cells (MFCs) employ anode microorganisms to catalytically degrade organic matter and directly convert chemical energy into electrical

energy through electron transfer and electrochemical reactions, achieving pollutant degradation while simultaneously recovering energy. This provides a completely new technological pathway for antibiotic wastewater treatment [9]. In recent years, MFCs have achieved a wealth of research results in the field of antibiotic removal, spanning from standalone MFC systems to their coupling with technologies such as constructed wetlands and photocatalysis, from the degradation of single antibiotics to synergistic removal under co-substrate conditions with multiple antibiotics, and from the elucidation of degradation mechanisms to the control of antibiotic resistance gene risks. Both the breadth and depth of research have been continuously expanding [10]. This review aims to systematically summarize the research progress of MFC-based antibiotic wastewater treatment and to provide a reference for further in-depth research and practical application of MFCs and their coupled technologies in treating antibiotic wastewater.

Introduction to and principles of microbial fuel cells

An MFC is a bio electrochemical system that directly converts chemical energy into electrical energy. Its core principle relies on the catalytic activity of anode electroactive microorganisms to efficiently transform the energy stored in organic matter into electricity. The structure of a typical dual-chamber MFC consists of an anode chamber, a cathode chamber, and a proton exchange membrane that separates the two [11,12]. The working process can be described in the following steps: (1) Substrate oxidation: The exaelectrons in the anode chamber degrade organic substrates (such as glucose, acetate, or organic matter in wastewater), releasing electrons and protons. This process is essentially microbial respiratory metabolism; the key distinction is that the terminal electron acceptor is not oxygen or a natural substance, but the anode. (2) Electron transfer: Microorganisms transfer electrons to the anode surface through various mechanisms. After being collected by the anode, the electrons flow through an external circuit to the cathode, performing work in the process. (3) Proton migration: Protons generated from substrate oxidation migrate through the solution and pass through the proton exchange membrane into the cathode chamber. (4) Cathodic reduction: The electrons arriving at the cathode combine with the migrated protons and an electron acceptor to form reduced products.

For an MFC using glucose as the substrate and oxygen as the electron acceptor, the overall reaction can be expressed as follows [13]:

Anode reaction: $C_6H_{12}O_6 + 6H_2O \rightarrow 6CO_2 + 24H^+ + 24e^-$

Cathode reaction: $6O_2 + 24H^+ + 24e^- \rightarrow 12H_2O$

Overall reaction: $C_6H_{12}O_6 + 6O_2 \rightarrow 6CO_2 + 6H_2O$

How microorganisms transfer electrons generated from intracellular metabolism to the extracellular electrode surface is the central scientific question of MFCs. Depending on the transfer pathway, extracellular electron transfer can be classified into two types: direct electron transfer and mediated electron transfer [14].

Direct electron transfer (DET) refers to the process in which microorganisms transfer electrons directly to the electrode through structural proteins on the cell membrane or pili that are in physical contact with the electrode. The most representative microorganisms include *Geobacter* and *Shewanella* species, along with the cytochrome c protein family. These proteins contain heme prosthetic groups that serve as electron carriers, enabling transmembrane electron transfer from the intracellular respiratory chain to the extracellular electrode. Some microorganisms can also form conductive pili known as “nanowires”, which enable long-distance electron transfer [15].

Mediated electron transfer (MET) relies on redox mediators secreted by the microorganisms themselves or added exogenously to complete electron transfer. These mediators can be reduced by the microorganisms in their reduced state, diffuse to the electrode surface where they are oxidized and transfer the electrons to the electrode, and then diffuse back to the microorganisms, forming a cycle. Common natural mediators include riboflavin and pyocyanin. Studies have shown that some microorganisms can simultaneously utilize both direct and mediated transfer pathways to achieve highly efficient extracellular electron transfer [16].

Research progress on the degradation of various antibiotics by MFCs

Antibiotics can be classified, based on their mechanisms of action, into β-lactams that inhibit cell wall synthesis, polymyxins that increase cell membrane permeability,

tetracyclines, chloramphenicol's, and aminoglycosides that interfere with protein synthesis, and quinolones and sulfonamides that inhibit nucleic acid replication. The degradation behavior of different antibiotic classes in MFCs and their effects on MFC performance differ significantly.

MFC degradation of antibiotics targeting the cell wall and cell membrane

(1) MFC degradation of β-lactam antibiotics

β-lactam antibiotics (e.g., penicillin, ceftriaxone sodium, and ampicillin) exert their antibacterial effects by inhibiting bacterial cell wall synthesis and represent the most widely used class of antibiotics in clinical practice. Liu et al. investigated the feasibility of treating penicillin-containing synthetic wastewater with an MFC. At a penicillin concentration of 50 mg/L and a glucose concentration of 1 g/L, the penicillin degradation efficiency reached 98.0%, while the maximum power density of the MFC reached 101.2 W/m³, which was six times higher than that of the control [17]. The same research group further examined the degradation of ceftriaxone sodium in an MFC. The results showed that at 50 mg/L ceftriaxone sodium, the removal efficiency reached 91.0% and the maximum power density was 113 W/m³ [18]. Other β-lactam antibiotics, such as ampicillin and cefazolin sodium, have also achieved degradation efficiencies exceeding 90.0% in MFCs, with the maximum power output substantially increased compared with those of groups without antibiotic addition [19,20]. These results indicate that β-lactam antibiotics can not only be metabolized and decomposed by anodic microorganisms as substrates, but their cell wall-disrupting mechanism may also reduce the apparent internal resistance of transmembrane electron transfer, thereby enhancing the power output of the MFC.

(2) MFC degradation of polypeptide antibiotics

Polymyxins are polypeptide antibiotics that act by binding to phosphate groups on the bacterial cell membrane, increasing membrane permeability, which leads to the leakage of intracellular small molecules and eventual cell lysis and death [21]. Yang et al. studied the effect of polymyxin on an *Rhodococcus pyridinivorans*-inoculated anode MFC [22]. Scanning electron microscopy observations revealed that after the addition of 10 mg/L polymyxin, pores appeared on the surface of the anode biofilm and the biomass decreased to some extent. However, the apparent internal resistance of the cell also decreased, and the maximum power output increased by 13.0%, reaching 68.8 W/m². This suggests that the membrane-damaging effect of polymyxin reduces the resistance of electron transmembrane transport to a certain degree, thereby simultaneously enhancing antibiotic degradation and electricity generation performance.

MFC degradation of antibiotics targeting protein synthesis and nucleic acid replication

(1) MFC degradation of quinolone antibiotics

Quinolone antibiotics (e.g., norfloxacin, ciprofloxacin, and enrofloxacin) kill bacteria by inhibiting DNA gyrase or topoisomerase IV, thereby blocking nucleic acid replication. Due to their potential effects on bone development, the environmental residues of these antibiotics have attracted considerable concern.

Ondon et al. investigated the degradation of norfloxacin and the removal of antibiotic resistance genes (ARGs) in MFC. At a norfloxacin concentration of 128 mg/L, the MFC achieved a maximum power density of 700 mW/cm² and a norfloxacin removal efficiency of 65.5%. More importantly, ARGs such as MDTK and MDTM in the MFC effluent were simultaneously eliminated, indicating that MFCs can not only degrade antibiotics but also reduce the risk of resistance gene dissemination [23]. Yan et al. treated ciprofloxacin with an MFC acclimated using swine farm wastewater. The optimal concentration was 10 mg/L, yielding a maximum output voltage of 600 mV and a degradation efficiency of 99.0% [24].

(2) MFC degradation of aminoglycoside antibiotics

Neomycin is an aminoglycoside antibiotic that interferes with protein synthesis by binding to the 30S ribosomal subunit. Ghanam et al. used an MFC acclimated with municipal wastewater to treat neomycin, achieving a degradation efficiency of 61.0% and a maximum power density of 140 mW/m² [25].

(3) MFC degradation of sulfonamide antibiotics

Sulfonamide antibiotics inhibit nucleic acid synthesis by blocking dihydrofolate synthase and thereby interfering with folate metabolism. Jiang et al. treated sulfadiazine in an MFC and achieved a removal efficiency as high as 97.3% [26]. Wen et al. treated sulfamethoxazole in a constructed wetland-coupled MFC system, attaining a removal efficiency of 87.5% and a power density of 1186.2 mW/m² [27]. Al-Ansari et al. used an MFC constructed with *Bacillus subtilis* to treat sulfanilamide, achieving a removal efficiency of 99.5% [28]. Studies have shown that the optimal degradation concentration

of sulfonamide antibiotics in MFCs is generally lower than those of quinolones and β-lactams, which may be related to the relatively low resistance level of sulfonamide-resistant bacteria.

In summary, the degradation behavior of different antibiotic classes in MFCs and their effects on MFC performance exhibit significant differences. The results are summarized in Table 1.

Table 1. Treatment effects of MFC on different types of antibiotics.

Antibiotic class	Name	Anode microbial source	Optimal concentration (mg/L)	Maximum power density	Removal rate (%)	References
β-Lactams	Penicillin	Effluent from another MFC	50.00	101.200 W/m^3	98.00	[17]
	Ceftriaxone sodium	Effluent from another MFC	50.00	113.00 W/m^3	91.00	[18]
	Cefazolin sodium	Aerobics activated sludge	200.00	30.400±2.100 mW/m^2	90.00	[19]
	Ampicillin	*Shewanel a putrefaciens*	10.00	1.956±0.015 mW/m^2	95.90±3.00	[20]
Polypeptides	Polymyxin B sulfate	*Rhodococcuspyridi nivorans*	10.00	68.800 W/m^2	/	[21]
Quinolones	Norfloxacin	Wastewater treatment plant	128.00	700.000 mW/cm^2	65.50	[23]
	Ciprofloxacin	Swine excreta	10.00	600.000 mV*	99.00	[24]
Aminoglycosides	Neomycin	Wastewater treatment plant	75.00	140.000±19.000 mW/m^2	61.00±7.00	[25]
Sulfonamides	Sulfadiazine	Anaerobic digester wastewater	0.35	128.000 mV*	97.30±2.70	[26]
	Sulfamethoxazole	Wastewater treatment plant	20.00	1186.200 mW/m^2	87.52±1.97	[27]
	Sulfanilamide	Anaerobic sludge	0.30	583.600 mV*	99.53	[28]

Note: * in the table indicates that power density data were not available in the cited literature, and the voltage is presented instead.

Research progress on the treatment of antibiotic wastewater by different biological processes

In recent years, the problem of antibiotic residues in wastewater has attracted widespread attention. Conventional wastewater treatment plants (WWTPs) are not specifically designed to remove antibiotics, and thus their removal efficiency is limited. This section mainly summarizes the degradation of antibiotic wastewater by different biological treatment processes and advanced treatment processes.

Conventional biological wastewater treatment processes

(1) Conventional activated sludge

Conventional activated sludge (CAS) is the most widely applied technology in biological wastewater treatment, and its antibiotic removal efficiency fluctuates greatly, ranging from negative removal to over 90.0%. Antibiotics are removed primarily through adsorption, especially for tetracyclines, fluoroquinolones, and macrolides. Studies have shown that extending the hydraulic retention time (HRT) and sludge retention time

(SRT) can significantly improve antibiotic removal efficiency. For example, after extending the HRT, the removal efficiency of sulfamethoxazole (SMX) increased from 45.0% to 80.0%. When the SRT was extended from 20 to 50 days, the removal efficiency of clarithromycin increased from less than 10.0% to over 90.0%.

(2) Sequencing batch reactor

Compared with CAS, sequencing batch reactors (SBRs) offer many advantages, such as simple structure, operational flexibility, and no need for an additional sludge recirculation system [29]. Studies have found that sulfonamides can be efficiently removed at SRT > 15 days and HRT > 4-6 h. Furthermore, anaerobic SBRs can maintain relatively good removal performance under high concentrations of sulfamethoxazole, although excessively high concentrations can inhibit microbial activity and biogas production [30].

(3) Anaerobic digestion

Anaerobic digestion (AD) is widely applied to treat organic wastewater containing high concentrations of COD and large amounts of antibiotics, and its antibiotic removal efficiency is superior to that of CAS [31]. Temperature is a key factor affecting AD performance, with removal efficiencies in summer significantly higher than those in winter [32]. In addition, the presence of antibiotics may inhibit methanogenic activity and reduce biogas production. Studies have shown that two-stage AD or the addition of iron oxides can alleviate the inhibitory effects, thereby improving antibiotic removal and methane production [33].

Advanced biological wastewater treatment processes

(1) Membrane bioreactor

Membrane bioreactors (MBRs) have attracted extensive attention in wastewater treatment in recent years. They combine adsorption, biodegradation, and membrane separation processes and offer advantages such as long SRT, low sludge production, and high microbial concentrations. In a laboratory-scale anaerobic membrane bioreactor, the removal efficiencies of trimethoprim (TMP) and sulfamethoxazole (SMX) through biodegradation reached 94.2% and 67.8%, respectively. Extending the SRT helps enrich slow-growing bacteria and improve degradation efficiency. However, membrane fouling is a major challenge for MBRs, especially when treating antibiotic-containing wastewater, which may alter sludge characteristics and exacerbate membrane fouling [34-37].

(2) Biological aerated filter system

Biologically aerated filter (BAF) systems have been applied to antibiotic wastewater treatment owing to their advantages of high organic loading, low cost, and low energy consumption. Studies have shown that at an initial concentration of 1 mg/L, the removal efficiency of ciprofloxacin (CIP) exceeded 95.0%. HRT is a key parameter affecting BAF performance; when the HRT was reduced from 10 h to 5 h, the CIP removal efficiency decreased from 95.0% to 79.0% [38]. Similar to MBRs, BAFs also suffer from clogging problems, which affect long-term operational stability.

(3) Microbial fuel cell system

MFC systems are an emerging technology applied for organic pollutant removal and energy recovery [39]. They require no additional reducing agents at the cathode or from an external power source and can achieve very high antibiotic removal rates. Compared with the 58.3% antibiotic removal in microbial control, the removal efficiency of oxytetracycline (OTC) in the MFC anode exceeded 99.0% [40]. The removal mechanisms include degradation of antibiotics as electron donors by anodic microorganisms, reductive degradation of antibiotics as electron acceptors at the cathode, and degradation through free radical attack. MFCs can also avoid the production of highly toxic byproducts from biological pathogens. For example, 3-amino-5-methylisoxazole, a highly toxic intermediate formed during SMX removal, was effectively degraded in the MFC [41].

Operating conditions such as applied voltage, electrode materials, and carbon source type significantly affect the MFC removal efficiency. When the applied voltage was 0.2 V, the degradation efficiency of nitrofurazone (NFZ) was 42.3%; when the applied voltage was increased to 0.8 V, the degradation efficiency of NFZ jumped to 70.6%. Electrode materials are fundamental to MFC performance and influence antibiotic removal. As biocatalysts, microorganisms enriched on the electrode stimulate electron transfer and antibiotic removal. Many researchers have focused on carbon materials as electrode materials. Carbon sources play a crucial role in microbial metabolic activities. For example, the removal

efficiency of NFZ with $NaHCO_3$ was slightly lower than that with glucose [42-44].

(4) Constructed wetland

Constructed wetlands (CWs) are typical environmentally friendly treatment technologies, mainly consisting of plants, substrates, and microorganisms. They remove antibiotics through adsorption, plant uptake, and microbial degradation [45]. Based on water flow patterns, CWs can be classified into free water surface (FWS), horizontal subsurface flow (HSSF), and vertical subsurface flow (VSSF) wetlands. VSSF exhibits better antibiotic removal performance than HSSF and FWS systems. For example, Liu et al. showed that the removal efficiencies of oxytetracycline (OTC) and CIP in VSSF were 98.0% and 97.0%, respectively, which were higher than those of 88.0% and 85.0% in FWS and 96.0% and 94.0% in HSSF [46]. The antibiotic removal performance in CWs is significantly influenced by multiple key factors, including flow direction, substrate type, plant species, and temperature [47].

Research progress on MFC systems coupled with other technologies for treating antibiotic wastewater

Although stand-alone MFC systems have demonstrated certain potential in antibiotic wastewater treatment, they still face limitations such as low degradation efficiency, inhibition of microbial activity, and restricted power output when treating highly persistent antibiotics or complex wastewater matrices. To overcome these limitations, researchers have coupled MFCs with other wastewater treatment technologies to construct various synergistically enhanced systems, significantly improving antibiotic removal efficiency and system stability. Currently, the most extensively studied coupled systems mainly include constructed wetland-MFC (CW-MFC), algae-MFC (A-MFC), bio-electro-Fenton MFC (BEF-MFC), sediment MFC (SMFC), and photocatalytic MFC (PMFC).

CW-MFC

Numerous studies have shown that MFCs can be integrated with CWs, where the anode and cathode electrodes are embedded into the CW, and the photosynthesis, root oxygen release, and microbial synergistic interactions of wetland plants create natural anaerobic (anode) and aerobic (cathode) zones [48]. Studies have demonstrated that CW-MFC can achieve a sulfamethoxazole removal efficiency of 82.4%, which is higher than the 71.3% obtained with a conventional constructed wetland [49]. Furthermore, the application of iron-carbon composite electrode materials or manganese ore electrodes can further enhance the system's electricity generation performance and antibiotic removal capability. For example, an iron-carbon enhanced CW-MFC achieved a ciprofloxacin removal efficiency as high as 91.2% and a maximum power density superior to that of an ordinary CW-MFC; it was also found to reduce the accumulation of antibiotics on the cathode, thereby decreasing the abundance of ARGs [50]. Direct reduction of iron can promote the electrochemical synthesis of ferrate, which in turn oxidizes antibiotics [51].

A-MFC

A-MFC is an advanced bioelectrochemical technology that utilizes photosynthesis in microalgal cells to supply oxygen to the cathode while simultaneously achieving carbon fixation and antibiotic degradation. Research has found that in an algae-bacteria coupled with MFC, the degradation efficiency of florfenicol can reach 79.6%, and the ammonia nitrogen removal efficiency approaches 100.0%. At initial concentrations below 20 mg/L, oxytetracycline can be almost completely removed within 48 hours [52].

BEF-MFC

BEF-MFC achieves efficient mineralization of recalcitrant antibiotics by generating H_2O_2 in situ at the cathode, which then reacts with Fe^{2+} to produce strongly oxidizing ·OH. Studies have shown that the removal efficiency of sulfamethoxazole by BEF-MFC was enhanced from 56.3% to 94.7% after cathodic treatment, and the removal efficiency of erythromycin could reach 88.7%. Enhancing the Fe^{2+}/Fe^{3+} cycle can further increase H_2O_2 production and antibiotic removal efficiency [53-55]. Although this system shows remarkable effectiveness in treating high-concentration antibiotic wastewater, challenges such as pH sensitivity and the generation of iron sludge still exist.

SMFC

SMFCs utilize organic matter in water body sediments as the anodic substrate, making them suitable for the in-situ remediation of antibiotics in natural water bodies.

Research has shown that SMFCs achieved an 86.2% removal efficiency of sulfamethoxazole within 20 days, and when combined with aquatic plants, the removal efficiency of antibiotic resistance genes exceeded 94.0%. In addition, a polyaniline/manganese dioxide modified anode significantly increased the removal efficiency of enrofloxacin (59.5%) and the maximum power density (165.09 mW/m²). SMFCs can also be coupled with an electro-Fenton system to form an SMFC-E-Fenton system, which can enhance the antibiotic removal efficiency to over 96.0% [56,57].

PMFC

PMFC introduces photocatalytic materials (such as TiO_2 and g-C_3N_4) to the cathode or anode, utilizing photogenerated electron-hole pairs to enhance redox reactions and thus improve antibiotic degradation efficiency [58]. A study found that a PMFC employing a TiO_2/GO/g-C_3N_4 heterojunction photoanode achieved tetracycline degradation efficiencies of 94.9% (anode) and 96.3% (cathode), with a maximum power density of 546 mW/m² [59]. A $LiNbO_3$/CF photocatalytic cathode achieved an ofloxacin removal efficiency of 86.5% at a concentration of 0.1 mmol/L, with a power density of 0.546 W/m² [60]. PMFCs have demonstrated excellent synergistic effects in the treatment of recalcitrant antibiotics, but issues such as the high cost of photocatalysts and low light utilization efficiency still need to be addressed.

Conclusion

Although MFCs and their coupled systems have made significant progress in treating antibiotic wastewater, they still face numerous challenges before large-scale applications. Future efforts should focus on the screening and directed domestication of highly tolerant electroactive functional consortia. This aims to improve the efficiency of antibiotic degradation and extracellular electron transfer. It is also necessary to explore low-cost electrode materials with high specific surface area and good biocompatibility. Such materials can strengthen microbial adhesion and electron transfer efficiency. Moreover, it is essential to optimize the operating parameters of coupled processes and solve key problems including high catalyst cost and membrane fouling. In addition, further studies should assess the removal mechanisms and potential residual risks of ARGs and ARB during MFC treatment. Correspondingly, coupled disinfection or advanced oxidation units should be developed to thoroughly prevent the spread of resistance. Accelerating the transition of MFC technology from laboratory scale to engineering applications in practical scenarios such as pharmaceutical wastewater and wastewater treatment plants will promote it as a low-carbon, sustainable antibiotic pollution control technology.

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Conflicts of Interests

The authors declare no conflict of interest.

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